

Three-dimensional Negative-Refractive-Index Metamaterials Based on All-Dielectric Coated Spheres

Na Fu, Weixing Shu,* Hailu Luo, Zhixiang Tang,

Yanhong Zou, Shuangchun Wen, and Dianyuan Fan

Key Laboratory for Micro/Nano Optoelectronic Devices of Ministry of Education,

School of Computer and Communications,

Hunan University, Changsha 410082, China

Abstract

A type of 3-dimensional optical negative-refractive-index metamaterials composed of all dielectric nanospheres is proposed and demonstrated theoretically. The metamaterials are constructed by pairing together two kinds of dielectric nanospheres as concentric shells embedded in a host medium. Mie-based extended effective theory shows that the dielectric core and the dielectric shell provide the negative permeability and the negative permittivity, respectively, both due to the strong Mie resonances. Within the coupled resonant frequency region, the negative index of refraction can be achieved.

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*Corresponding author. *E-mail address:* wxshuz@gmail.com.

I. INTRODUCTION

Negative-index metamaterials(NIMs) [1] are artificial structures that exhibit negative index of refraction. Since these metamaterials enable a variety of novel applications such as superlens [2], optical nanocircuits [3], cloaking [4, 5], marvellous progresses on the NIMs have been made [6–8] in recent years.

Along with the rapid development of NIMs, it has been challenging to design 3D isotropic NIMs at optical frequencies. So far, efforts devoted to designing NIMs at optical frequencies mainly consist of two basic ideas, namely those based on L-C resonant models and the Mie scattering models. The most prominent structures based on L-C models are metal-dielectric fishnet structures [9]. However, due to the anisotropy of these metallic structures, the negative refraction can only be performed in one certain direction. Another drawback of the metal based structures is the difficulty in enhancing resonant frequencies and fabrication. The Mie scattering model based structures are those multilayered metal and dielectric microspheres[10], metal coated dielectric spheres [11], et al. Also, there are some problems, such as large losses and saturation effect, inherent in metals at optical frequencies associated with these metallic metamaterials. As an alternative, the Mie resonance of highly polaritonic dielectric materials, such as rod [12–14] or cube type [15] metamaterials, provides a more promising way to design low-loss, much simpler 3D metamaterials with higher frequencies. However, the ultimate goal of realizing 3D metamaterials at optical frequencies has not been fulfilled up to now.

In this paper, we theoretically propose a type of optical negative refractive index metamaterial that composed of all dielectric coated nanospheres. Our objective is to use low-permittivity dielectric materials to increase the electromagnetic resonant frequencies to optical domain, and to utilize highly symmetrical sphere-type structure to realize 3D optical NIMs. For dielectric nanospheres, a strong magnetic dipole resonance results in the negative effective permeability. And an electric dipole resonance leads to the negative effective permittivity when the dielectric constant and radius of the spheres are increased. We proceed as follows. Firstly, we use Mie theory to describe the effective magnetic and electric resonances, and then derive the relationship between the effective permeability and permittivity and the material parameters by homogenizing the spherical scatters. Secondly, we show that the negative magnetic and electric response can be produced at optical frequencies by tuning

the sphere parameters. Lastly, we tune these resonances together using coated dielectric spheres and the negative refractive index can be obtained. Theoretically, a negative index of refraction can be obtained at any optical frequency.

II. NEGATIVE MAGNETIC AND ELECTRIC RESPONSE BY MIE-BASED MAXWELL-GARNETT THEORY

The theoretical basis of our study is the Mie-based Maxwell-Garnett(MMG) theory [16]. We consider a composite of small dielectric spheres of radius a and dielectric constant ϵ_r embedded in a host medium with dielectric constant ϵ_h and incident wavelength λ_h . Generally Mie scattering can be used when $(0.05 \sim 0.1)\lambda_h < 2a < (3 \sim 6)\lambda_h$ [17]. Let x be the size parameter which can be defined as $x = 2\pi a/\lambda_h$. If $x \ll 1$, for a periodic distribution, the effective permeability μ_{eff} is given by Clausius-Mossotti equation,

$$\frac{\mu_r^{eff} - 1}{\mu_r^{eff} + 2} = \frac{f}{a^3}\alpha, \quad (1)$$

where α is the particle dipole polarizability, $f = 4\pi N a^3/3$ is the filling fraction of the composite, and N is the number density of the spheres.

As long as x is small enough so that the Mie coefficients a_m and b_m with $m > 1$ can be neglected, a size dependent extension of the MG formula [18] suggests that in terms of the Mie coefficient the polarizability is

$$\alpha = i \frac{3a^3}{2x^3} b_1, \quad (2)$$

and the Mie scattering coefficients, a_m and b_m are

$$a_m = \frac{n\psi_m(nx)\psi'_m(x) - \psi'_m(nx)\psi_m(x)}{n\psi_m(nx)\xi'_m(x) - \psi'_m(nx)\xi_m(x)}, \quad (3)$$

$$b_m = \frac{\psi_m(nx)\psi'_m(x) - n\psi'_m(nx)\psi_m(x)}{\psi_m(nx)\xi'_m(x) - n\psi'_m(nx)\xi_m(x)}, \quad (4)$$

respectively, where $\psi_m(x)$ and $\xi_m(x)$ are related to the Riccati-Bessel functions.

Combined with Eqs. (1) and (2), the effective permeability for a homogenizing distribution of inclusions can be obtained as

$$\mu_r^{eff} = \epsilon_h \left(1 + \frac{6\pi i N_\mu b_1}{\epsilon_h^3 k^3 - 2\pi i N_\mu b_1} \right), \quad (5)$$

where $k = \omega_\mu^{inc}/c$, and ω_μ^{inc} is the frequency of the incident wave. Analogously, the effective permittivity ϵ_r^{eff} can be expressed as

$$\epsilon_r^{eff} = \epsilon_h \left(1 + \frac{6\pi i N_\epsilon a_1}{\epsilon_h^3 k^3 - 2\pi i N_\epsilon a_1} \right). \quad (6)$$

Eqs. (5) and (6) imply that the effective permeability and permittivity depend on the frequency of the incident wave, the host medium, Mie coefficient as well as the number density of the spheres.

A. Magnetic response

Eq. (5) reveals the magnetic resonant frequency ω_μ^{res} occurs if the frequency of the incident wave satisfy

$$\omega_\mu^{inc} = \frac{c}{\epsilon_h} (2\pi i N_\mu b_1)^{1/3}. \quad (7)$$

Eq. (7) implies the magnetic response requires appreciable values of the $N_\mu b_1$. For a moderate filling fraction, the magnetic resonance mainly results from the fundamental Mie resonance [14]. The scattering properties of the sphere-type structures may therefore be understood by studying the resonant behavior of b_1 . Using the half-integer Bessel function, $\psi_m(x) = \sqrt{\pi x/2} J_{m+1/2}(x)$ and $\xi_m(x) = \sqrt{\pi x/2} [J_{m+1/2}(x) + (-1)^m J_{-m-1/2}(x)]$, for $m = 1$, the simplified resonance occurs when $J_0(nka) = \sin(nka)/(nka) = 0$ [11]. The fundamental Mie resonance frequency is $\omega_\mu^{res} = \pi c / (a \sqrt{\epsilon_r \epsilon_h})$. We assume, as is typical for dielectric resonators, that the resonance frequencies are determined by the real part of the refractive index of the sphere $n = \sqrt{\epsilon_r}$. The relative permittivity of polaritonic dielectrics follows $\epsilon_r = \epsilon_\infty [1 + (\omega_L^2 - \omega_T^2) / (\omega_T^2 - \omega^2 - i\omega\gamma)]$ where ϵ_∞ is the high frequency limit of the dielectric permittivity, γ is the loss factor, and ω_T and ω_L are the transverse and longitudinal optical phonon frequencies. Since the electromagnetic resonant frequency region we considered is much larger than ω_T , so in this paper we take the approximation of $\epsilon_r = \epsilon_\infty$.

Theoretically, almost arbitrary values of μ_{eff} can be obtained in a collection of appropriate size of non-magnetic spheres. As an example, consider a collection of dielectric spheres using the parameters: $\epsilon_r = n^2 = 2.2$ with the number density $N_\mu = (242.6nm)^{-3}$ and radius $a = 120nm$. For simplicity, we choose host medium as $\epsilon_h = 1.0$. According to Eq. (7), the magnetic resonance is predicted at 310THz. A full calculation of Eq. (5)(shown in Fig. 1)

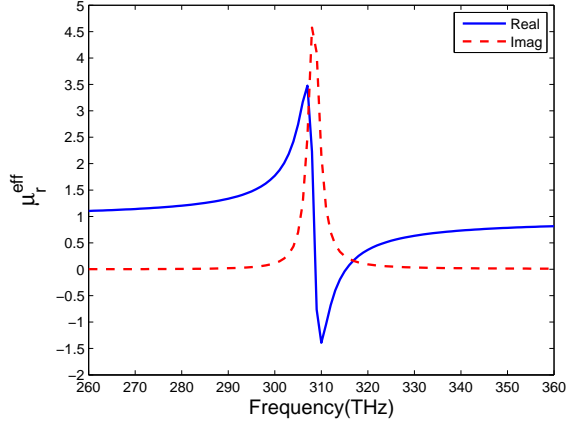


FIG. 1: Calculated effective permeability μ_{eff} for a periodic distribution of dielectric spherical particles with radius $a = 120nm$ and the dielectric constant $\epsilon_r = 2.2$. The dielectric constant of the host medium is $\epsilon_h = 1.0$ and the number density of the composite is $N_\mu = (242.6nm)^{-3}$.

reveals a resonance in μ_{eff} centered at 310THz with a real value of -1.5, which agrees well with our prediction. Additionally, from Eq. (7) we also find the number density of spheres N_μ and the dielectric constant of the host medium ϵ_h will affect the magnetic resonant frequency. This is vigorous to assist the design of a negative refractive index in Sec. III.

B. Electric response

The method described in deciding the magnetic response can also be used to calculate a_1 . From Eq. (6), the dielectric resonance ω_ϵ^{res} is induced when

$$\omega_\epsilon^{inc} = \frac{c}{\epsilon_h} (2\pi i N_\epsilon a_1)^{1/3}. \quad (8)$$

The resonant frequencies of a_1 in the long wavelength limit can be estimated by

$$\frac{J_0(nka) + J_2(nka)}{J_0(nka) - J_2(nka)} + \frac{1}{\epsilon_r} = 0. \quad (9)$$

Eq. (9) is only suitable for numerical calculation and the resonant frequency is related to the dielectric constant of the nanospheres ϵ_r . Since the first Mie resonance a_1 occurs in lower frequencies than the fundamental Mie resonance b_1 with the same parameters, it requires a dielectric constant larger than that in Sec. II A to drive an equivalent resonance.

Take the parameter $\epsilon_r = 13.4$ of LiTO_3 [19], and the same number density as in the above section. The lattices of LiTO_3 spheres are predicted to possess the fundamental dielectric

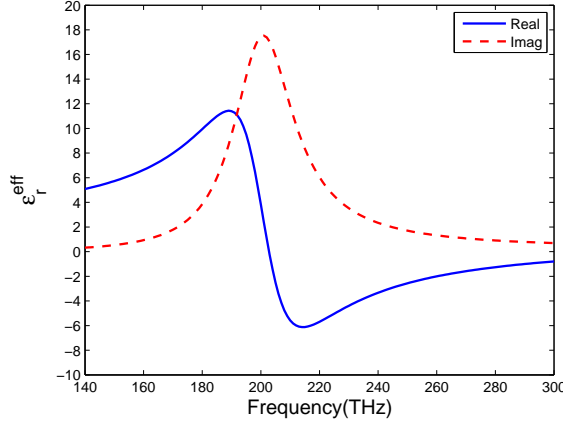


FIG. 2: Calculated effective permeability ϵ_{eff} for a periodic distribution of identical dielectric spherical particles with radius $a = 120nm$ and the dielectric constant $\epsilon_r = 13.4$. The dielectric constant of the host medium is $\epsilon_h = 1.0$ and the number density is $N_\mu = (242.6nm)^{-3}$.

response at 212THz. Calculation of Eq. (6) (shown in Fig. 2) indicates that the first electric resonance is induced at approximately 210THz with a value of -7. This is particularly helpful to provide a desired electric response in Sec. III.

III. NEGATIVE INDEX OF REFRACTION

In the above two section, we have realized the negative permeability and the negative permittivity separately. However, a single type of dielectric spheres collection cannot serve as a negative refractive index metamaterial on its own. In order to pair together the magnetic resonant frequency and the electric resonant frequency in the same frequency region, one can require $N_\mu b_1 = N_\epsilon a_1$. In particular, we find that utilizing the relationship between size parameter x and the Mie resonant frequency can narrow the gap between a_1 and b_1 . Concentric dielectric nanospheres enable the problem to be solved and provide a novel mechanism for the creation of 3D negative index metamaterial. We can choose dielectric sphere as the core to provide a desired magnetic resonance, and larger radius shell with higher dielectric constant material cover on the core to increase the electric resonant frequencies to corresponding region. When these two resonances simultaneously exist, the negative refractive index can be obtained. Detailed computational analysis is as follows.

According to the regulation of the Mie scattering coefficient, we consider a collection of

coated dielectric composite particles, with the core (region 1) designed as Sec.II A ($a_1 = 120nm, \epsilon_1 = 2.2$) which provides $\mu_r^{eff} < 0$, and with the shell (region 2) designed as Sec.II B ($a_2 = 130nm, \epsilon_2 = 13.4$) which provides $\epsilon_r^{eff} < 0$, embedded in a host medium (region 3, $\epsilon_3 = 1.0$). The particle number density is fixed to $N_\epsilon = N_\mu = (271.4nm)^{-3}$. This can be considered as a periodic lattice of nanospheres with radius $a = 130nm$ and periodicity $R = 480nm$. Scattering of Electromagnetic waves from coated spheres has been worked out and a_1, b_1 are written as [20, 21]

$$a_1 = \frac{\psi_1(x_2) - s_1\psi_1'(x_2)}{\xi(x_2) - s_1\xi_1'(x_2)}, \quad (10)$$

$$b_1 = \frac{t_1\psi_1(x_2) - \psi_1'(x_2)}{t_1\xi_1(x_2) - \xi_1'(x_2)}, \quad (11)$$

respectively, where

$$s_1 = n_2 \frac{\psi_1(n_2x_2) - p_1\chi_1(n_2x_2)}{\psi_1'(n_2x_2) - p_1\chi_1'(n_2x_2)}, \quad (12)$$

$$t_1 = n_2 \frac{\psi_1'(n_2x_2) - q_1\chi_1'(n_2x_2)}{\psi_1(n_2x_2) - q_1\chi_1(n_2x_2)}, \quad (13)$$

$$p_1 = \frac{n_2\psi_1(n_2x_2)\psi_1'(n_1x_1) - n_1\psi_1'(n_2x_2)\psi_m(n_1x_1)}{n_2\psi_1'(n_1x_1)\chi_1(n_2x_2) - n_1\psi_1(n_1x_1)\chi_1'(n_2x_2)}, \quad (14)$$

$$q_1 = \frac{n_2\psi_1'(n_2x_2)\psi_1(n_1x_1) - n_1\psi_1'(n_1x_1)\psi_1(n_2x_2)}{n_2\psi_1(n_1x_1)\chi_1'(n_2x_2) - n_1\psi_1'(n_1x_1)\chi_1(n_2x_2)}. \quad (15)$$

Here $x_1 = k_0r_1$, $x_2 = k_0r_2$ and $\chi_1(z) = -zy_1(z)$ where $y_1(z)$ is the spherical Bessel function of the second kind. With the parameters stated above, magnetic resonant frequency is predicted at 265Thz and dielectric resonant frequency is predicted at 250THz by substituting a_1 and b_1 in Eq. (7) and Eq. (8). To get the effective material values for coated spheres, we simply substitute a_1 and b_1 in Eqs. (5) and (6). The full calculations of the effective permeability and permittivity are shown in Fig. 3. These results coincide with our prediction. The effective index is calculated with $n_{eff} = \sqrt{\mu_r^{eff}\epsilon_r^{eff}}$ and ensuring $n_{eff}'' \geq 0$. The maximum negative index is obtained with the value about -1.2 at 260Thz where the magnetic and the dielectric response simultaneously occur.

In the above, a periodic distribution of coated nanospheres is shown to exhibit negative refractive index at optical frequencies. As for random distribution of units, previous studies

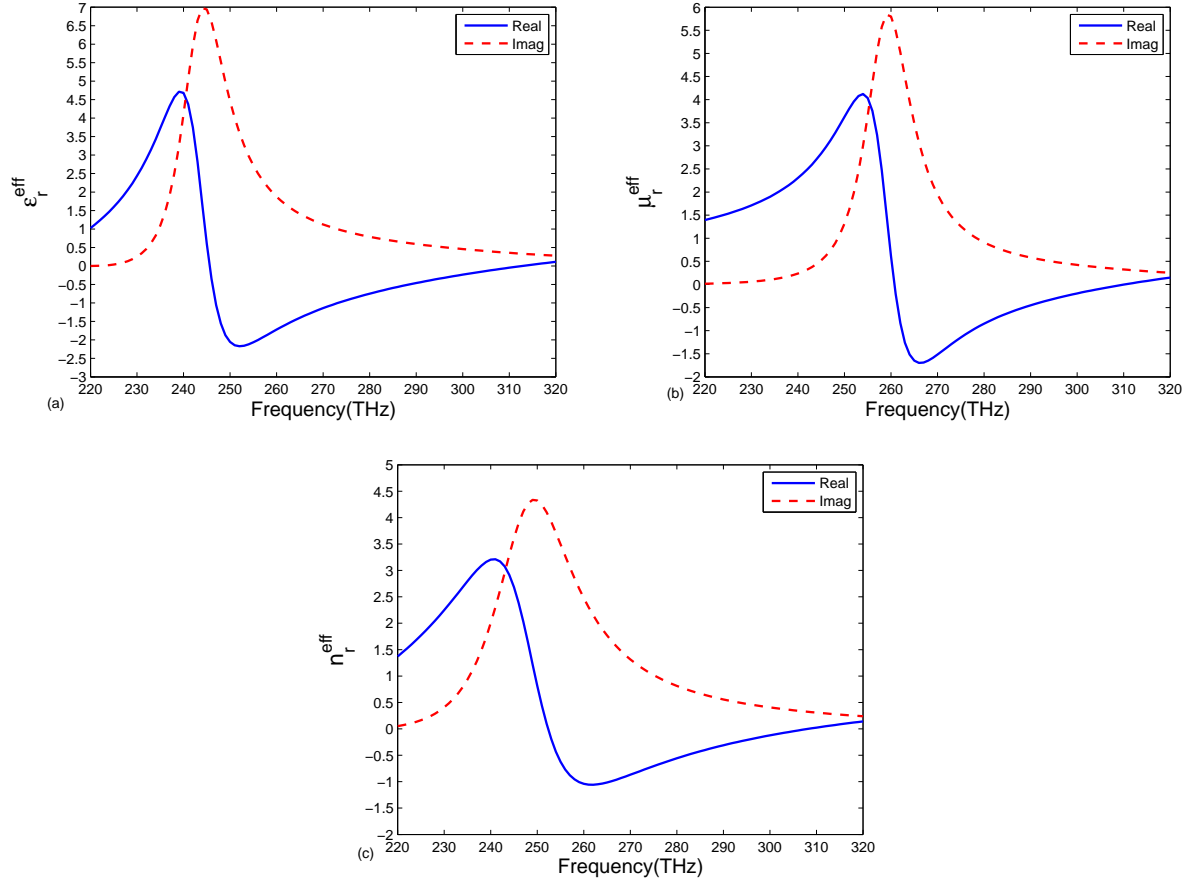


FIG. 3: The effective permittivity, permeability and refractive index of a collection of coated spheres. The dielectric constant of the core is $\epsilon_1 = 2.2$ and the radius $a_1 = 120nm$, and the coating is made of $LiTO_3$ with $\epsilon_2 = 13.4$ and $a_2 = 130nm$. The dielectric constant of the host medium is $\epsilon_3 = 1.0$ and the number density are $N_\epsilon = N_\mu = (271.4nm)^{-3}$.

have shown that the effective magnetic and electric resonances are different from periodic structure. Specifically, Yannopapas has proven that disordered distribution of elements can slightly affect values of μ_r^{eff} and ϵ_r^{eff} . However, the magnetic and electric resonances behave qualitatively in the same way as in the periodic case [22, 23]. It is because, though the symmetry of the lattice can slightly influence the Mie resonance, the determinant factors are the size, number density and dielectric properties of the spheres (as shown in Eqs. (5) and (6)) [11, 19, 24]. Therefore, even nonperiodic distribution of coated spheres stated in this paper can realize the NIMs at optical frequencies.

IV. CONCLUSION

In this paper we have theoretically shown a type of all dielectric NIM at optical frequency by properly designing related parameters of dielectric nanospheres. Near the frequencies of magnetic and electric Mie resonances provided by the dielectric core and shell respectively, both negative permeability and negative permittivity are produced, and then left-handed metamaterials are obtained in the optical domain. Note that LiTO_3 coated nanosphere is just taken as demonstration. Actually, due to the highly tunable parameters of the all-dielectric concentric spheres, these proposed structures enable a broad frequency range of negative refractive index from deep infrared to visible domain. This is important if such structures are to be used in practical NIM-based applications at optical frequencies. Future work will be desirable to choose appropriate dielectric materials for the coated spheres to fabricate 3D isotropic negative refractive index metamaterial in visible frequencies.

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